

# Improvement of magnetic hardness at finite temperatures: *ab initio* disordered local moment approach for YCo<sub>5</sub>

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Temperature dependence of the magnetocrystalline anisotropy energy and magnetization of the prototypical rare-earth magnet YCo<sub>5</sub> is calculated from first principles, utilizing the relativistic disordered local moment approach. We discuss a strategy to enhance the finite-temperature anisotropy field by hole doping, paving the way for an improvement of the coercivity near room temperature or higher.

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## I. INTRODUCTION

Ferromagnetism in permanent magnets has been a long-standing problem in solid state physics and materials science. The mechanism of coercivity, resistance of the spontaneous magnetization against the externally applied magnetic field in the reverse-direction, is not yet entirely known after almost 70 years since Brown's paradox<sup>1</sup>. Development of modern rare-earth magnets like Nd<sub>2</sub>Fe<sub>14</sub>B<sup>2</sup> and SmCo<sub>5</sub><sup>3,4</sup> triggered material-specific investigations of the coercivity mechanism<sup>5</sup>. They culminate in today's state-of-the-art technological optimization of the microstructure of ferromagnetic materials for the enhanced coercivity<sup>6</sup> on the phenomenological and empirical basis. More fundamental understanding and controlling on the basis of solid-state physics and statistical physics is desired to better use these technological advances. Especially the temperature decay of coercivity is a problem in industrial applications and thus more and more demands for the control of finite-temperature properties of permanent magnets are rising.

The high-temperature tail of the magnetism is mostly carried by 3*d*-electrons<sup>7</sup> in the intermetallic system between transition-metal (TM) and rare-earth (RE) elements while the low-temperature magnetism is dominated by 4*f*-electrons. Thus developing an understanding and the theoretical control over the temperature decay of 3*d*-electron magnetism has the universal utility for the optimization of high-temperature magnetism of RE magnets. Thus we focus on YCo<sub>5</sub> as our target system, extracting the 3*d*-electron part out of the SmCo<sub>5</sub> family. The so-called 1-5 magnets represent the RE magnets with one of the simplest crystal structures and SmCo<sub>5</sub>-based magnets are 2<sup>nd</sup> in magnetic performance only to the champion-magnet family including Nd<sub>2</sub>Fe<sub>14</sub>B; SmCo<sub>5</sub> currently serves for some special applications under extreme conditions<sup>8</sup> with its high Curie temperature and stability against corrosion. Also the large magnetocrystalline anisotropy energy (MAE) of 5.5 [MJ/m<sup>3</sup>] at room temperature  $T = 295$  [K] in YCo<sub>5</sub><sup>9</sup> can let itself qualify for permanent magnet applications<sup>8</sup>.

Describing the magnetic properties of such a material presents significant challenges to *ab initio* theory. Itinerant electron magnetism in realistic magnetic materials has often been addressed in the ground state with the application of the density-functional theory (DFT). Incorporation of finite temperature physics and description of correlated electron behavior can be issues for DFT which does not always include localized electron effects adequately into its scope. Both issues require a combination of DFT and statistical physics in some way. Here we follow the idea of disordered local moments<sup>10–13</sup> to incorporate the physics of thermal fluctuations of well-developed magnetic moments into the spin-polarized density functional theory (DFT) for ferromagnetic materials<sup>14–17</sup>. A relativistic version of the disordered local moment (DLM) approach has recently been developed<sup>18,19</sup> and successfully applied to finite-temperature magnetic anisotropy in L1<sub>0</sub>-FePt alloys<sup>18,19</sup> and Co films<sup>20</sup> where magnetism is all carried by *d*-electrons. Thus the *d*-electron part in the magnetism of rare-earth magnets can be a good target for DLM.

In this work we present *ab initio* calculations based on DLM for YCo<sub>5</sub> at finite temperatures. From calculated temperature dependence of the magnetization  $M$  and the MAE, which we denote by  $K_{u1}$  as it mostly comes from the uni-axial magneto-crystalline anisotropy (MCA) for YCo<sub>5</sub>, we address the thermal decay of the anisotropy field estimated as  $H_A = 2K_{u1}/M$  which generally shows the approximate proportionality to the coercivity field  $H_c$ <sup>21</sup>. The relation is practically  $H_c \sim 0.2H_A$  with the practical hard upper limit being  $H_c \lesssim 0.5H_A$  that is reached only in some exceptional samples<sup>22</sup>. Assuming such rough proportionality, we propose a strategy to improve  $H_A(T)$  at high temperatures in the range  $T \sim 400 - 500$  [K]. Our main message is encapsulated in Fig. 1 which shows that hole-doped YCo<sub>5</sub> with the number of doped holes being 0.24 and 0.35 per unit cell has the stronger anisotropy field than pure YCo<sub>5</sub> in the temperature range  $T \gtrsim 450$  [K] and  $T \gtrsim 550$  [K], respectively.

Experimentally, doping-enhanced coercivity has been

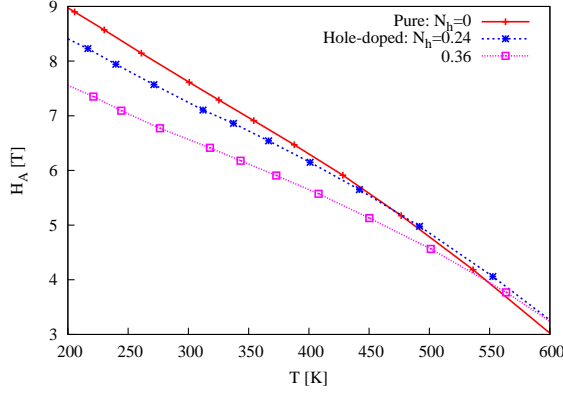
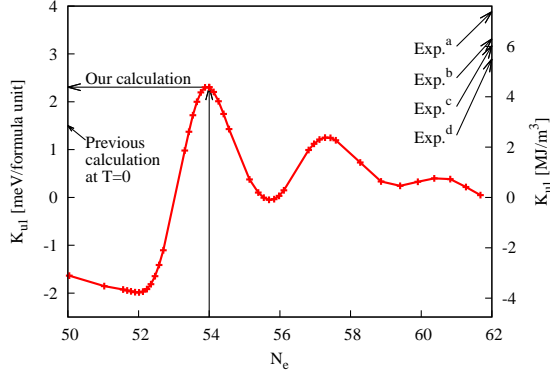


FIG. 1. (Color online) Calculated temperature dependence of the anisotropy field  $H_A$  for pure and hole-doped  $\text{YCo}_5$ , where doping-induced enhancement of  $H_A$  is observed in the temperature range  $T \gtrsim 450$  [K] and 550 [K] with the number of doped holes being  $N_h = 0.24$  and 0.36, respectively.



<sup>a</sup>  $K_{u1} = 7.38$  [MJ/m<sup>3</sup>] at  $T = 4.2$  [K] from Ref. 23.

<sup>b</sup>  $K_{u1} = 6.3$  [MJ/m<sup>3</sup>] at  $T = 77$  [K] from Ref. 24.

<sup>c</sup>  $K = 6.03$  [MJ/m<sup>3</sup>] from Ref. 25 at  $T = 293$  [K]. We assume  $K_{u1}$  dominates in  $K$  as was shown in Ref. 23.

<sup>d</sup>  $K_{u1} = 5.5$  [MJ/m<sup>3</sup>] at room temperature from Ref. 8.

FIG. 2. (Color online) Calculated magnetic anisotropy energy plotted as the function of the valence electron number for  $\text{YCo}_5$  near the ground state  $T = 0$  [K]. The numbers on the left-hand side of the vertical axis indicates the results per the formula unit of  $\text{YCo}_5$  and those on the right-hand side of the vertical axis gives the same quantities converted to per the unit volume using the experimental lattice constants in Ref. 26. The previous theoretical result for MAE at  $T = 0$  is taken from Ref. 27. The number of valence electrons per formula unit of  $\text{YCo}_5$  is 54.

known for  $\text{YCo}_{5-x}\text{Ni}_x$  and other  $\text{RECo}_{5-x}\text{Cu}_x$  magnets ( $\text{RE}=\text{Ce}, \text{Sm}$ )<sup>28</sup>. The coercivity is indeed significantly affected by doping, which is seen in our calculated results on the filling dependence of MAE near the ground state as shown in Fig. 2. On top of such a filling dependence of coercivity determined by the electronic struc-

ture, our idea is to let the electronic states below the peak of MAE in Fig. 2 be thermally populated so that the high-temperature MAE is enhanced. Experimentally observed doping-enhanced coercivity had been discussed in conjunction with intrinsic pinning<sup>28</sup>. Our results on the finite-temperature physics together with the electron band filling-sensitive nature of MAE point to a new scenario for doping-enhanced coercivity at high temperatures.

The rest of paper is organized as follows. Our proposal on the strategy to enhance the high-temperature coercivity is found in Sec. IV. This follows an outline of the DLM methodology underpinning the calculations in Sec. II and Sec. III. Conclusions are given in Sec. V.

## II. METHODS

Our computational method, the DLM approach, is based on the Korringa-Kohn-Rostoker (KKR) method<sup>29,30</sup> and coherent potential approximation (CPA)<sup>31</sup> for *ab initio* electronic structure calculations. Especially to address the temperature dependence of MAE which is brought about by spin-orbit interaction (SOI), we follow the relativistic formulation<sup>18,19</sup>. These aspects in Sec. II A and Sec. II B, describe the general framework and how to extract MAE, respectively at the level of sketching the basic ideas. The MAE as our key observable is focused on and for the rest of the observables that we present in Sec. III, we refer to the original works<sup>15,19</sup> and a recent review article<sup>32</sup> for their explicit expressions in terms of KKR-CPA. In Sec II C we give the details of the application to the case of  $\text{YCo}_5$ .

### A. Framework

#### 1. Separation of fast and slow dynamics

Under the situation that spin-fluctuation dynamics are much slower than the characteristic time scales of electronic motions<sup>15</sup>, well developed classical local moments  $\hat{\mathbf{e}}$  are assumed to exist on each site  $i = 1 \dots N$  in the unit cell, with  $N$  being the number of magnetic atoms in the unit cell. The thermal average  $\langle \rangle$  of the local moment

$$\langle \hat{\mathbf{e}}_i \rangle = \int \dots \int \hat{\mathbf{e}}_i P^{(\hat{n})}(\{\hat{\mathbf{e}}\}) d\hat{\mathbf{e}}_1 \dots d\hat{\mathbf{e}}_N \quad (1)$$

is assumed to be aligned with the magnetization direction  $\hat{\mathbf{n}}$  for a ferromagnet. Here  $\hat{\mathbf{n}}$  is fixed in the calculation by hand. We have specified the configuration of the local moments by  $\{\hat{\mathbf{e}}\}$  and statistical weight  $P^{(\hat{n})}(\{\hat{\mathbf{e}}\})$  given in terms of the Boltzmann weights at finite temperatures.  $\langle \hat{\mathbf{e}}_i \rangle = \mathbf{m}_i$  is a magnetic order parameter. For a ferromagnet, magnetized along a direction  $\hat{\mathbf{n}}$ ,  $\mathbf{m}_i = m_i \hat{\mathbf{n}}$ .

## 2. DLM as a realistic mean field theory

DLM is implemented as a realistic mean-field theory for the local moments embedded in a sea of electrons given by spin-polarized DFT for the target material. For a given configuration of the local moments

$$\{\hat{\mathbf{e}}\} \equiv \{\hat{\mathbf{e}}_1, \hat{\mathbf{e}}_2, \dots, \hat{\mathbf{e}}_N\},$$

the Boltzmann weight is defined as

$$\exp \left[ -\beta \Omega^{(\hat{\mathbf{n}})}(\{\hat{\mathbf{e}}\}) \right],$$

referring to the grand potential  $\Omega^{(\hat{\mathbf{n}})}(\{\hat{\mathbf{e}}\})$  in the spin-resolved density functional theory for a given target ferromagnet ( $\beta = 1/k_B T$ ). The partition function is accordingly written as

$$Z^{(\hat{\mathbf{n}})} = \int \dots \int \exp \left[ -\beta \Omega^{(\hat{\mathbf{n}})}(\{\hat{\mathbf{e}}\}) \right] d\hat{\mathbf{e}}_1 \dots d\hat{\mathbf{e}}_N$$

and the probability distribution function (PDF) in Eq. (1) of the local-moment configuration  $\{\hat{\mathbf{e}}\}$  is

$$P^{(\hat{\mathbf{n}})} = \frac{\exp \left[ -\beta \Omega^{(\hat{\mathbf{n}})}(\{\hat{\mathbf{e}}\}) \right]}{Z^{(\hat{\mathbf{n}})}}$$

Here we use the mean-field approximation (MFA)

$$\Omega_0^{(\hat{\mathbf{n}})}(\{\hat{\mathbf{e}}\}) \stackrel{\text{MFA}}{=} \sum_{i=1}^N \mathbf{h}_i^{(\hat{\mathbf{n}})} \cdot \hat{\mathbf{e}}_i \quad (2)$$

where the Weiss field  $\mathbf{h}_i^{(\hat{\mathbf{n}})} = h_i^{(\hat{\mathbf{n}})} \hat{\mathbf{n}}$  is written as follows<sup>15</sup>.

$$h_i^{(\hat{\mathbf{n}})} = \int \frac{3}{4\pi} (\hat{\mathbf{e}}_i \cdot \hat{\mathbf{n}}) \left\langle \Omega^{(\hat{\mathbf{n}})} \right\rangle_{\hat{\mathbf{e}}_i} d\hat{\mathbf{e}}_i \quad (3)$$

for  $i = 1 \dots N$  where  $i$  runs over the local moments in the unit cell and the thermal average denoted by  $\langle \cdot \rangle_{\hat{\mathbf{e}}_i}$  implies the restriction in the averaging process with the direction of the local moment on site  $i$  fixed to that specified by  $\hat{\mathbf{e}}_i$ . (Further terms proportional to, say,  $(\hat{\mathbf{e}}_i \cdot \hat{\mathbf{n}})^2$ , can be added to Eq. 2 to improve the mean field description<sup>41</sup> but have a rather small effect.) With the MFA in Eq. (2) the overall partition function factorizes into contributions from each of local moments

$$Z^{(\hat{\mathbf{n}})} = \prod_{i=1}^N Z_i^{(\hat{\mathbf{n}})}$$

and the partition function is written in terms of the Weiss field

$$\begin{aligned} Z_i^{(\hat{\mathbf{n}})} &= \int \exp \left( -\beta \mathbf{h}_i^{(\hat{\mathbf{n}})} \cdot \hat{\mathbf{e}}_i \right) d\hat{\mathbf{e}}_i \\ &= \frac{4\pi}{\beta h_i^{(\hat{\mathbf{n}})}} \sinh \beta h_i^{(\hat{\mathbf{n}})} \end{aligned} \quad (4)$$

and the PDF is written as follows:

$$\begin{aligned} P_i^{(\hat{\mathbf{n}})}(\hat{\mathbf{e}}_i) &= \exp \left( -\beta \mathbf{h}_i^{(\hat{\mathbf{n}})} \cdot \hat{\mathbf{e}}_i \right) / Z_i^{(\hat{\mathbf{n}})} \\ &= \frac{\beta h_i^{(\hat{\mathbf{n}})}}{4\pi \sinh \beta h_i^{(\hat{\mathbf{n}})}} \exp \left( -\beta \mathbf{h}_i^{(\hat{\mathbf{n}})} \cdot \hat{\mathbf{e}}_i \right) \end{aligned} \quad (5)$$

With this expression for the PDF, the free energy is written as follows:

$$\begin{aligned} F^{(\hat{\mathbf{n}})} &= \left\langle \Omega^{(\hat{\mathbf{n}})} \right\rangle \\ &+ \frac{1}{\beta} \sum_{i=1}^N \int P_i^{(\hat{\mathbf{n}})}(\hat{\mathbf{e}}_i) \log P_i^{(\hat{\mathbf{n}})}(\hat{\mathbf{e}}_i) d\hat{\mathbf{e}}_i \end{aligned} \quad (6)$$

where the first term describes the internal energy and the second describes  $-TS$  with the magnetic entropy

$$S = - \sum_{i=1}^N \int P_i^{(\hat{\mathbf{n}})}(\hat{\mathbf{e}}_i) \log P_i^{(\hat{\mathbf{n}})}(\hat{\mathbf{e}}_i) d\hat{\mathbf{e}}_i.$$

The magnetization is given using the PDF in Eq. (5) as follows.

$$\mathbf{m}_i^{(\hat{\mathbf{n}})} = \langle \hat{\mathbf{e}}_i \rangle \equiv m_i^{(\hat{\mathbf{n}})} \mathbf{h}_i^{(\hat{\mathbf{n}})} \quad (7)$$

$$m_i^{(\hat{\mathbf{n}})} = -\frac{1}{\beta h_i^{(\hat{\mathbf{n}})}} + \coth \beta h_i^{(\hat{\mathbf{n}})} = L(\beta h_i^{(\hat{\mathbf{n}})}) \quad (8)$$

Here  $L(x)$  is the Langevin function.

## 3. CPA to embed the thermal disorder into DFT

Coherent Potential Approximation (CPA)<sup>31</sup> is an approach to deal with the disorder physics on a mean-field level by averaging over the random potentials to introduce an effective uniformly distributed potential. Following the original idea by Hasegawa<sup>11</sup> and Hubbard<sup>12</sup>, thermal disorder in local moments is embedded into the description of the electronic states<sup>15</sup> on the level of CPA.

The way that the CPA determines the effective medium is by letting the motion of an electron simulate the motion of an electron on the average. The scattering problem at the heart of KKR is solved to get the single-site  $t$ -matrix  $\underline{t}_i$  for each local moment  $i$  where the underlined  $t$ -matrix is that in orbital and spin angular momentum space. Some more elaboration on that solution is given below with Eq. (12). For the system magnetized along the direction  $\hat{\mathbf{n}}$ , the medium is specified by a set of CPA-imposed  $t$ -matrices<sup>33</sup>

$$\underline{t}_c^{(\hat{\mathbf{n}})} \equiv (\underline{t}_{1,c}, \underline{t}_{2,c}, \dots, \underline{t}_{N,c})$$

which is determined (for details we refer to Ref. 19) for a given set of Weiss fields  $h_i^{(\hat{\mathbf{n}})}$  as in Eq. (3) and corresponding  $P_i^{(\hat{\mathbf{n}})}(\hat{\mathbf{e}}_i)$  as in Eq. (5), where the finite-temperature physics is encapsulated.

The  $t$ -matrices  $\underline{t}_c^{(\hat{\mathbf{n}})}$  that specify the effective medium comes into the formulation of DFT as follows. By magnetic force theorem we consider only the single-particle

energy part of the spin-resolved DFT grand potential as our effective local-moment Hamiltonian

$$\Omega^{(\hat{n})}(\{\hat{\mathbf{e}}\}) \simeq - \int dE f_{\text{FD}}(E; \mu^{(\hat{n})}) N^{(\hat{n})}(E; \{\hat{\mathbf{e}}\}), \quad (9)$$

where  $f_{\text{FD}}(E; \mu^{(\hat{n})})$  is the Fermi-Dirac distribution function with the chemical potential being  $\mu^{(\hat{n})}$  and  $N^{(\hat{n})}(E; \{\hat{\mathbf{e}}\})$  is the integrated density of states (DOS)<sup>19,34</sup> with the given direction of magnetization  $\hat{n}$  and the given configuration of local moments  $\{\hat{\mathbf{e}}\}$ . Using the Lloyd formula<sup>35</sup> for  $N^{(\hat{n})}(E; \{\hat{\mathbf{e}}\})$  on the right-hand side of Eq. (9), the grand potential is expressed in terms of the  $t$ -matrices<sup>19</sup>.

## B. Calculation of magnetic anisotropy energy

### 1. Defining the MAE for the uni-axial MCA

For uniaxial magnets, the free energy takes the form

$$F^{(\hat{n})} = F_{\text{iso}} + K_{\text{u1}} \sin^2 \theta$$

where  $F_{\text{iso}}$  is the isotropic part,  $K_{\text{u1}} > 0$  is the MAE for uni-axial MCA and  $\theta$  is the angle between the direction of magnetization and the easy axis. Then

$$\begin{aligned} T_{\theta} &= - \frac{\partial F}{\partial \theta} \\ &= -2K_{\text{u1}} \sin \theta \cos \theta \end{aligned} \quad (10)$$

Thus in order to extract MAE for the uni-axial MCA we fix the direction of magnetization to be  $\hat{n} = (1, 0, 1)/\sqrt{2}$  that is,  $\hat{n} = (\sin \theta \cos \phi, \sin \theta \sin \phi, \cos \theta)$  with  $\theta = \pi/4$  and  $\phi = 0$  and calculate the magnetic torque  $T_{\theta=\pi/4}$ . Our target MAE is obtained as

$$K_{\text{u1}} = -T_{\theta=\pi/4}. \quad (11)$$

### 2. Spin-Orbit Interaction (SOI) in relativistic KKR

For a given set of self-consistent potentials, electronic charge, and local moment magnitudes, the directions of local moments are described by the unitary transformation of the single-site  $t$ -matrix for local moment  $i$

$$\underline{t}_i(E; \hat{\mathbf{e}}_i) = \underline{R}(\hat{\mathbf{e}}_i) \underline{t}_i(E; \hat{\mathbf{z}}) \underline{R}(\hat{\mathbf{e}}_i)^{\dagger} \quad (12)$$

where  $\underline{t}(E; \hat{\mathbf{z}})$  is the  $t$ -matrix with the effective field pointing along the local  $z$ -axis for given energy  $E$ , and  $\underline{R}(\hat{\mathbf{e}}_i)$  is for the  $O(3)$  unitary transformation that rotates the  $z$ -axis along the direction of  $\hat{\mathbf{e}}_i$ . The scattering problem at the heart of KKR is solved incorporating SOI with a magnetic field pointing along the  $z$ -axis<sup>36</sup> and the single-site  $t$ -matrix  $\underline{t}_i(E; \hat{\mathbf{z}})$  is obtained.

In the absence of SOI, an element of the single site scattering  $t$ -matrix

$$t_{i;l,m,\sigma;l',m',\sigma'} = t_{i;l,\sigma} \delta_{l,l'} \delta_{m,m'} \quad (13)$$

where  $l$  is the angular momentum quantum number and  $m$  the azimuthal component and

$$\underline{t}_i(E; \hat{\mathbf{e}}_i) = \underline{t}_i^+ \underline{1} + \underline{t}_i^- \sigma \cdot \mathbf{e}_i \quad (14)$$

where an element  $t_{l,m;l',m'}^{+(-)} = \frac{1}{2}(t_{i;l,\frac{1}{2}} + (-)t_{i;l,-\frac{1}{2}})$ . When SOI is included, the  $t$ -matrix is no longer independent of  $m$  which leads to the generation of magnetic anisotropic effects.

### 3. Torque-based formula in KKR-CPA

The uni-axial MAE is obtained as the derivative of the free energy following Eq. (10). Inserting Eq. (6) for the free energy, we get the following<sup>19</sup>.

$$T_{\theta}^{(\hat{n})} = - \frac{\partial}{\partial \theta} \sum_i \int P_i^{(\hat{n})}(\hat{\mathbf{e}}_i) \langle \Omega^{(\hat{n})} \rangle_{\hat{\mathbf{e}}_i} d\hat{\mathbf{e}}_i \quad (15)$$

This is our working formula to produce the results for the finite-temperature MAE of YCo<sub>5</sub> which we now go on to describe.

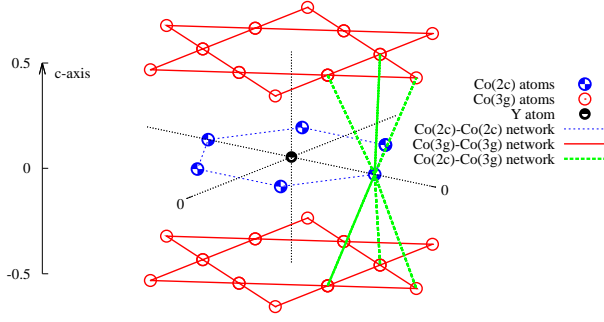
## C. Specifics with the case of YCo<sub>5</sub>

The past decade has seen the successful application of DLM theory to L1<sub>0</sub>-FePt alloy<sup>18,19</sup> which is a uniaxial ferromagnet and the magnetism is carried largely by 3d and 5d electrons. Since YCo<sub>5</sub> involves mostly 3d-electrons for its magnetism, success of DLM description on the same level as was achieved for L1<sub>0</sub>-FePt is expected provided that the assumption of localized moment on the magnetic atoms works well. Some care must be taken in the application of DLM for Co-based magnets since the trend among Fe, Co, and Ni<sup>13</sup> shows that the assumption of localized moment works well for Fe, faces challenge for Ni, and Co sits somewhere in-between. Stability of local moments on the Co sublattices in the present case is discussed in Sec. II C 2. Indeed we will see below some inherent underestimate near the Curie point in our calculations which might indicate some fragile nature of the local moments in YCo<sub>5</sub>. This problem can be cured by extending the calculation to that based on the non-local CPA<sup>37,38</sup>. For now we are mostly concerned with the lowest temperature properties and the middle-temperature range of  $T \lesssim 600$  [K] which is well below the Curie temperature at  $T_{\text{Curie}} = 920$  [K]<sup>26</sup>. So we proceed with the single-site theory within the scope of the present project.

### 1. Crystal structure

We take the experimental crystal structure of YCo<sub>5</sub> with the space group No. 191  $a = 4.948$  [Å] and  $c = 3.975$  [Å]<sup>26</sup>. The unit cell consists of one formula unit: Y, Co(2c), and Co(3g) sublattices. The atomic

(a)



(b)

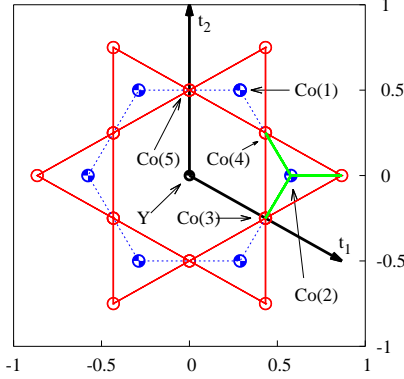


FIG. 3. (Color online) Crystal structure of  $\text{YCo}_5$ : (a) bird's eye view and (b) top view seen along the  $c$ -axis. Nearest-neighbor pairs in the  $\text{Co}(2c)$ - $\text{Co}(3g)$  network are only partially drawn for the illustration. Labels for atoms in (b) follow Table I.

configuration in the unit cell is shown in Fig. 3 (a) as a bird's eye view. The top view along the  $c$ -axis is shown in Fig. 3 (b) to illustrate the relative positions of the  $\text{Co}(2c)$ -atoms and the  $\text{Co}(3g)$ -atoms. Within the layer along the  $ab$ -plane, it is seen that nearest-neighbor  $\text{Co}(3g)$ -atoms form a kagomé lattice. Between those kagomé layers,  $\text{Co}(2c)$ -atoms sit right on the center of  $\text{Co}(3g)$ -triangles to form a hexagonal lattice on the same  $ab$ -plane layer as Y-atoms. The ratio  $c/a \simeq 0.80$  means that the tetrahedron formed by the  $\text{Co}(3g)$ -triangle and the  $\text{Co}(2c)$ -atom

sublattice	atom	atomic coordinates
Y		(0, 0, 0)
Co(2c)	Co(1)	( $1/(2\sqrt{3})$ , $1/2$ , 0)
	Co(2)	( $1/\sqrt{3}$ , 0, 0)
Co(3g)	Co(3)	( $\sqrt{3}/4$ , $-1/4$ , $1/2$ )
	Co(4)	( $\sqrt{3}/4$ , $1/4$ , $1/2$ )
	Co(5)	(0, $1/2$ , $1/2$ )
primitive translation vectors		
$\mathbf{t}_1$		( $\sqrt{3}/2$ , $-1/2$ , 0)
$\mathbf{t}_2$		(0, 1, 0)
$\mathbf{t}_3$		(0, 0, 1)

TABLE I. Set-up of the unit cell of  $\text{YCo}_5$  in the present calculations.

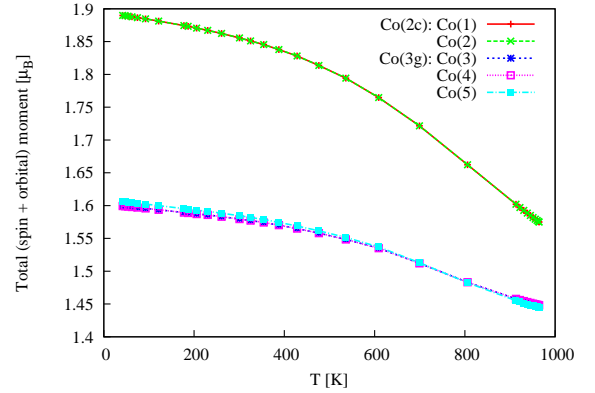


FIG. 4. (Color online) Calculated temperature dependence of the total (including both spin and orbital) local moment for each magnetic sublattice in  $\text{YCo}_5$ .

is close to the regular one. So the  $\text{Co}(2c)$ - $\text{Co}(3g)$  atomic distance is almost equal to  $\text{Co}(3g)$ - $\text{Co}(3g)$  distance which is  $0.5a$ . On the other hand,  $\text{Co}(2c)$ - $\text{Co}(2c)$  distance which is  $a/\sqrt{3} \simeq 0.58a$  spans the longest distance among the nearest-neighbor Co atom pairs. We set the position of each atom and the direction of the translation vectors as shown in Table I in our calculation. The labeling scheme for the atoms and the directions is illustrated in Fig. 3 (b).

## 2. Robustness of local moments in $\text{YCo}_5$

We inspect the validity of DLM approach at finite temperatures in the present calculation. Calculated temperature dependence of each of local moments in  $\text{YCo}_5$  are shown in Fig. 4. The DLM picture of magnetism in metals at finite temperature is based on the assumption that there are some aspects of the interacting electrons of a material that vary slowly in comparison with faster degrees of freedom. These are captured in terms of the orientational unit vectors,  $\{\hat{\mathbf{e}}\}$ , the transverse part of

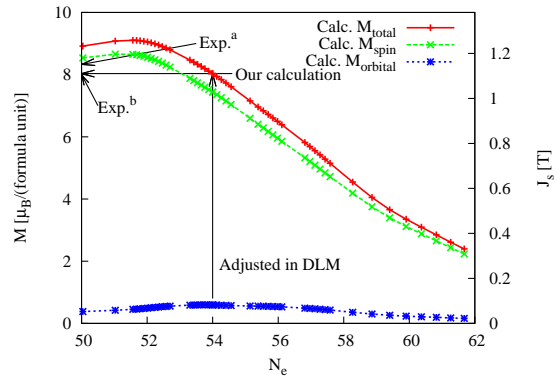
the magnetic fluctuations. The electronic grand potential  $\Omega^{(\mathbf{n})}(\{\hat{\mathbf{e}}\})$  is in principle available from spin-resolved DFT (SDFT) generalised for the non-collinear magnetic profiles which are labelled by  $\{\hat{\mathbf{e}}\}$ <sup>15</sup>. Within the tenets of SDFT the charge and magnetisation densities depend on these configurations, i.e.  $\rho(\mathbf{r}, \{\hat{\mathbf{e}}\})$ ,  $\vec{M}(\mathbf{r}, \{\hat{\mathbf{e}}\})$ . In the space around an atom at a site  $i$  the magnetisation is constrained to follow the orientation  $\hat{\mathbf{e}}_i$  there but its magnitude  $\mu(\mathbf{r})$  can depend on the surrounding orientational environment,  $\mu(\mathbf{r}) = \mu(\mathbf{r}, \{\hat{\mathbf{e}}\})$ . In many of the materials where the DLM theory has been successfully applied, the magnitudes are found to be rather insensitive to the environment and can be modelled accurately as rigid local moments. In other materials, e.g. Ni,<sup>39</sup> the magnetism is driven by the coupling of the magnitudes, (longitudinal magnetic fluctuations) with these transverse modes. When there is short-range aligning of the orientations or long range magnetic order, a finite magnetisation magnitude can establish; whereas in environments where the orientations differ significantly over short distances, the magnitudes shrink.

Figure 4 indicates the extent of this effect in YCo<sub>5</sub>. We have started with a SDFT calculation of the charge and magnetisation density  $\rho(\mathbf{r})$  and  $\vec{M}(\mathbf{r})$  for the  $T = 0$ K ferromagnetic state,  $\{\hat{\mathbf{e}} = \hat{\mathbf{n}}\}$  and find the magnetisation magnitude per site is 1.9 and 1.6  $\mu_B$  for the Co(2c) and Co(3g) sites respectively. When we use the scf potentials that these produce in a frozen potential approximation in our DLM theory for increasing temperatures when the moment orientations become disordered, the average magnetisation magnitude per site output from this calculation diminishes. For temperatures up to  $\sim 600$  [K], this decrease is slight, thereafter more significant as seen in the figures. Up to 600 [K] therefore the DLM theory as applied is adequate and we can examine the effects of transverse fluctuations on the MAE that it describes. For higher temperatures however, the effects of short-ranged correlations among the  $\hat{\mathbf{e}}$  orientation (via the use of the non-local CPA for example<sup>37,38</sup>) and longitudinal fluctuations<sup>40</sup> need to be addressed for a more complete picture. Comprehensive analyses on bcc-Fe, where the robustness of the local moments are established, can be found in a recent work<sup>41</sup>.

Further details about the calculations are described in Appendix A.

### III. RESULTS

We start with the calculated MAE near the ground state to demonstrate its sensitivity to the filling of the electrons in Fig. 2. Corresponding data for the magnetization is shown in Fig. 5. We see that even the sign of the MAE sensitively changes depending on a fraction in the filling of electrons, which is reasonable in the physics of magnetic anisotropy where adding/removing one electron in an electronic cloud that points to a particular real-space direction in a given crystal-field environment in-



<sup>a</sup>  $M = 8.33 [\mu_B]$  at liquid-helium temperature from Ref. 23.

<sup>b</sup>  $M = 7.99 [\mu_B]$  at room temperature from Ref. 26.

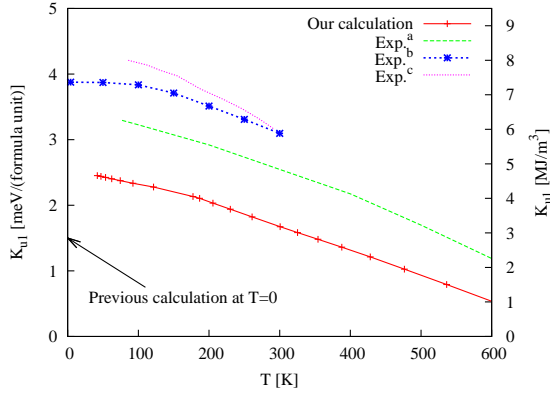
FIG. 5. (Color online) Calculated magnetization of YCo<sub>5</sub> plotted as a function of the number of electrons near the ground state at  $m = 0.967$ . Our calculated result 8.03  $[\mu_B]$  at around  $T = 100$  [K] falls onto the experimental numbers within 2 digits. For the closeness of such data to the ground state, see Fig. 7.

deed affect which direction the magnetic moment should prefer under the presence of SOL. So the challenge is how precisely we can pin-point the electron number right onto the realistic one, on which details are given in Sec. A 3. Fixing the electron number by a fine-tuning of the chemical potential in the DLM runs, we obtain the temperature dependence of MAE and magnetization. The temperature dependence of the anisotropy field is deduced from them. These data for the bulk are shown in Sec. III A. Then we inspect how our calculated MAE scales with respect to the calculated magnetization, resolving into sublattices and temperature ranges in Sec. III B. Comparison to the single ion anisotropy model analyses by Callen and Callen<sup>42</sup> and more recent ones<sup>18,43</sup> is carried out to help uncover the key mechanisms.

#### A. Bulk observables

Calculated temperature dependence of MAE for YCo<sub>5</sub> by DLM at the fixed correct valence-electron number is shown in Fig. 6. We have a systematic underestimate of MAE for the overall temperature range as compared to the experimental results found in the literature. However the qualitative temperature dependence is well reproduced. In the similar manner to MAE, calculated temperature dependence of magnetization at the fixed valence-band filling is shown in Fig. 7. Fitting the calculated temperature dependence of magnetization as it decreases to zero in the temperature range  $T > 900$  [K] to the following relation

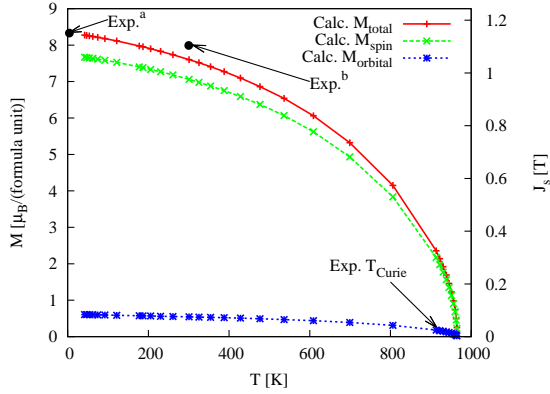
$$M(T) = A_M |T - T_{\text{Curie}}|^\beta,$$



<sup>a</sup> Ref. 24 for  $K_{u1}$ . <sup>b</sup> Ref. 23 for  $K_{u1}$ .

<sup>c</sup> Ref. 25 for  $K$ , with  $K_{u1}$  dominating.

FIG. 6. (Color online) Calculated temperature dependence of magnetic anisotropy energy for  $\text{YCo}_5$ . Previous calculation result at  $T = 0$  is taken from Ref. 27.



<sup>a</sup> Ref. 23 <sup>b</sup> Ref. 26

FIG. 7. (Color online) Calculated temperature dependence of magnetization for  $\text{YCo}_5$ . Experimental Curie temperature, 920 [K], is taken from Ref. 26.

we get  $T_{\text{Curie}} = 965$  [K],  $\beta = 0.50$ , and  $A_M = 0.33$ . The critical exponent falls onto the mean-field value  $\beta = 1/2$  within the numerical accuracy as it should, which gives a check of the data. Our *ab initio* result for the Curie temperature compares with the experimental Curie temperature  $T_{\text{Curie,exp}} = 920$  [K]<sup>26</sup> within the deviation of 5%. A mean-field result on the transition temperature typically gives an overestimate by  $O(10)\%$  as was observed for  $\text{L1}_0\text{-FePt}$  alloy calculated by exactly the same method<sup>18</sup> as the one we utilize here; the apparent absence of such an overestimate might rather indicate the fragility of the local moments close to  $T_{\text{Curie}}$ .

We obtain the *ab initio* result for the temperature dependence of the anisotropy field  $H_A$  as shown in Fig. 8 (a) following the simple coherent magnetization rotation pic-

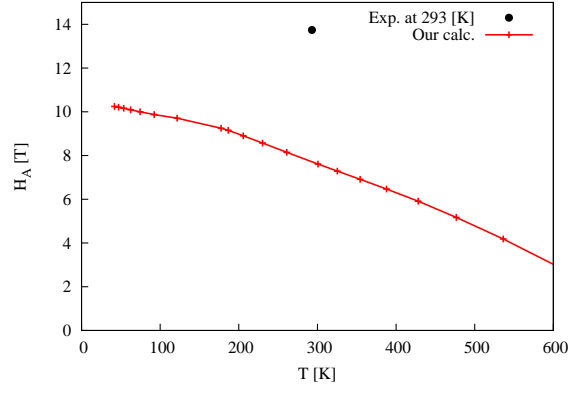


FIG. 8. (Color online) Temperature dependence of the anisotropy field for  $\text{YCo}_5$  as deduced from the numbers presented in Figs. 6 and 7, following Eq. (16). The experimental data at 293 [K] is taken from Ref. 25.

ture,

$$K_{u1} = \frac{1}{2} M H_A, \quad (16)$$

with  $K_{u1}$  and  $M$  being the calculated uniaxial MAE in Fig. 6 and magnetization in Fig. 7, respectively. The underestimate by 30% of  $K_{u1}$  on the left-hand side of Eq. (16) leads to a similar underestimate of  $H_A$ .

## B. Scaling MAE and Magnetization

Scaling relation concerning MAE with respect to magnetization

$$K_{u1}(T) = A [M(T)/M(0)]^\alpha \quad (17)$$

has been discussed for magnetic materials of which the representative one was established by Callen and Callen<sup>42</sup> on the basis of single-ion theory. Their exponent  $\alpha = l(l+1)/2$  at low temperatures, where  $l$  is the order of the expansion in terms of the spherical harmonics, has been challenged in some materials<sup>18,43</sup>. Callen-Callen argument for a ferromagnet with the uni-axial anisotropy predicts an exponent  $\alpha = 3$  with  $l = 2$ . Here we inspect what sort of scaling relation holds for our target magnet  $\text{YCo}_5$ .  $\text{YCo}_5$  is an intermetallic compound and the single-ion theory is not directly applicable to it. On top of that, the particular crystal structure of  $\text{YCo}_5$  comes with multiple magnetic sublattices in the unit cell. The bulk scaling between MAE and magnetization might not be exactly the same as sublattice-resolved scaling concerning the fact that each sublattice is exposed to its own crystal-field environment, depending on its own characteristic temperature dependence.

We start with the temperature dependence of local-moment resolved MAE as shown in Fig. 9. Exceptional



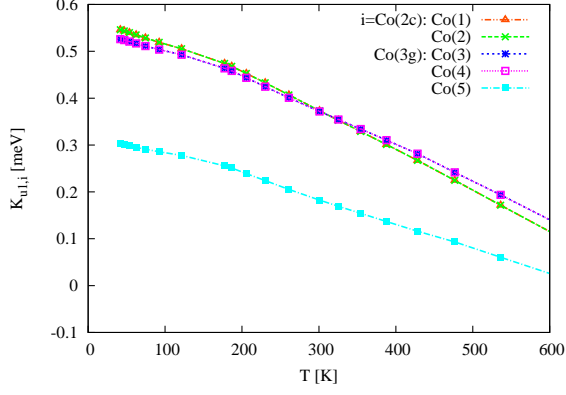


FIG. 9. (Color online) Calculated temperature dependence of local-moment-resolved MAE.

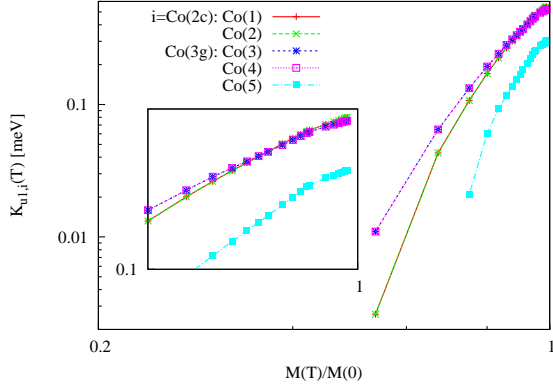


FIG. 10. (Color online) Calculated MAE plotted as a function of the order parameter,  $m \equiv M(T)/M(0)$ , for each local moment  $i$ . The inset is a zoomed-in plot in region  $M(T)/M(0) \lesssim 1$ .

behavior seen for Co(5) in the Co(3g)-sublattice originates in the effect of SOI with the magnetization direction  $\mathbf{n} = (1, 0, 1)$ . Out of the data in Fig. 9, the sublattice-resolved MAE as a function of the order parameter  $m = M(T)/M(0)$  is plotted in Fig. 10 to which we apply the scaling relation Eq. (17) for each of the contributions from the magnetic sublattices as well as for the bulk data. The bulk scaling analysis is shown representatively in Fig. 11. Experimental bulk data is taken from the past literature<sup>23</sup>. For the temperature range where the reliable experimental data set seems to be available, we tabulate the extracted scaling exponents in Table II. The scaling exponents do depend on the magnetic sublattices. We find that the bulk scaling exponent is observed only in a phenomenological way after summing up the contribution of each magnetic atoms in the unit cell. The overall calculated trend from the high-temperature side with larger  $\alpha$  to the low-temperature side with smaller  $\alpha$  is shared by all of the sublattice and the bulk. The bulk

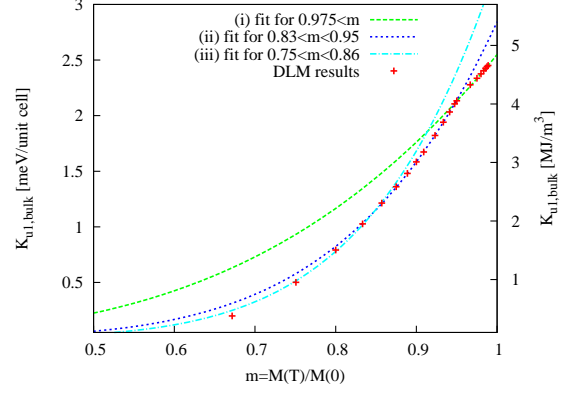


FIG. 11. (Color online) Scaling of the bulk MAE,  $K_{ul,bulk}(T)$ , with respect to the order parameter  $m = M(T)/M(0)$ . The scaling exponent  $\alpha$  in  $K_{ul,bulk}(T) = Am^\alpha$  depends on the temperature-dependent scaling region: i)  $\alpha = 3.5$  for  $m > 0.975$  which corresponds to 41 [K]  $< T < 75$  [K], ii)  $\alpha = 5.6$  for  $0.83 < m < 0.95$  which corresponds to 186 [K]  $< T < 477$  [K], and iii)  $\alpha = 6.5$  for  $0.75 < m < 0.86$  which corresponds to 428 [K]  $< T < 610$  [K].

	$T = 177$ [K]	186 [K]	300 [K]
Co(1)/Co(2)	–		$\alpha = 5.6$
Co(3)/Co(4)	–		$\alpha = 4.9$
Co(5)			$\alpha = 7.7$
Bulk (Our calc.)	–		$\alpha = 5.6$
Bulk (Exp. <sup>a</sup> )	$\alpha = 6.1$ for $150$ [K] $\leq T \leq 300$ [K]		

<sup>a</sup> Ref. 23

TABLE II. Focus temperature range and the extracted scaling exponents  $\alpha$  in  $K_{ul} \propto m^\alpha$  for each local moment on the magnetic sublattice in the unit cell of YCo<sub>5</sub> and for the bulk.

scaling exponent around the room temperature is calculated to be  $\alpha \sim 5.6$  which reasonably compares with the experimental exponent  $\alpha \sim 6.1$  that was extracted in the temperature region including the room temperature.

We note that our calculated data in Fig. 11 shifts from experimental data on the  $m$  axis when compared at the same temperatures. Our calculated data for  $m > 0.96$  are for below the liquid-nitrogen temperatures while for the experimental data<sup>23</sup> the room-temperature data is already in the parameter range  $m > 0.96$ . That deviation could be related to the inherent underestimate in our calculations.

#### IV. DISCUSSIONS

We discuss the strategy to improve the persistence of coercivity against temperature decay. It has been known that the coercivity is approximately proportional to the anisotropy field  $H_A$ <sup>5,21</sup>. A significant part of the reduc-



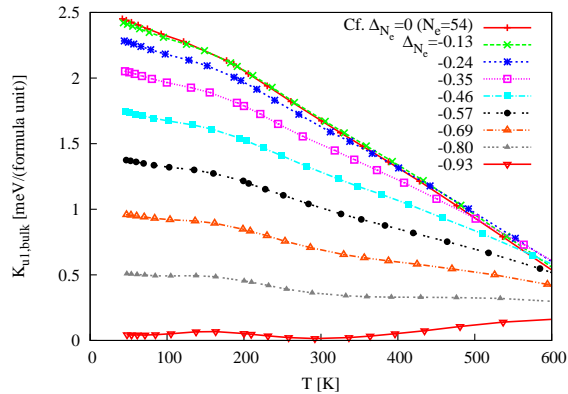


FIG. 12. (Color online) Calculated temperature-dependence of MAE for the hole-doped YCo<sub>5</sub>.

tion factor  $H_c/H_A$  originates from the microstructure which is beyond the scope of the bulk electronic structure considerations. Now our interest lies in how to improve  $H_A(T)$  or equivalently  $K_{u1}(T)$  for high  $T$ . We propose a scheme to exploit the peak position of MAE in Fig. 2 and how the electrons are thermally excited from the ground state following the Fermi-Dirac distribution. The idea is to let the thermally excited electrons populate the targeted electronic state which lies below the peak in Fig. 2 to enhance MCA. For that, we artificially lower the chemical potential to make some place for thermally excited electrons to populate at high temperatures. Then we sacrifice the optimal values of the MCA at low temperatures but we can extract the optimal anisotropy field over the operating temperature ranges.

The results for the calculated temperature dependence of the MAE in the hole-doped YCo<sub>5</sub>'s is shown in Fig. 12. Hole doping is implemented by artificially lowering the Fermi level in DLM runs for YCo<sub>5</sub>. Measuring the position of the lowered Fermi level,  $E_{F,\text{doped}}$ , from the undoped one,  $E_{F,\text{undoped}}$ , in units of Kelvins helps us to figure out around which temperature range the thermally excited electrons would reach the peak position in Fig. 2. We found that for the reduction range  $150 \text{ [K]} \lesssim \Delta E_F \lesssim 300 \text{ [K]}$ , with  $\Delta E_F \equiv (E_{F,\text{doped}} - E_{F,\text{undoped}})$ , some enhancement of the MAE around  $T \sim 500 \text{ [K]}$  is numerically observed. The data with  $\Delta E_F = 314 \text{ [K]}$  and  $471 \text{ [K]}$  were extracted with the corresponding reduction in the electron number being  $\Delta N_e = -0.24$  and  $-0.35$ , respectively, as the main message of this paper in Fig. 1, where MAE is expressed in terms of the anisotropy field following Eq. (16).

Remarkably, non-monotonic temperature dependence of  $H_A$  is found for the computational “sample” in the low-coercivity region with the electron number reduced by around 0.9 in Fig. 12. This non-monotonic temperature dependence of MAE is reminiscent of what has been known experimentally for Y<sub>2</sub>Fe<sub>14</sub>B since 1986, where the temperature-enhancement in MAE in the order of 0.2-0.3 [MJ/m<sup>3</sup>] is observed around  $T \sim 300 \text{ [K]}$ <sup>44</sup>.

The common factor among Y<sub>2</sub>Fe<sub>14</sub>B and YCo<sub>5</sub> is that  $3d$ -electron contribution in the MCA of rare-earth magnets is extracted out. The non-monotonicity in our finite-temperature data for the chemical-potential controlled YCo<sub>5</sub> comes from the following two factors: a) thermal population of the particular electronic states that corresponds to the peak of MAE in Fig. 2, and b) various contributions at finite temperatures from multiple sublattices in the unit cell. We speculate that analogous physics may be at work in Y<sub>2</sub>Fe<sub>14</sub>B which would require more extensive computations than those for the present project.

We note that the Curie temperature shifts as the chemical potential is reduced. The temperature dependence of magnetization shifts rather monotonically with respect to the artificially manipulated chemical potential while  $K_{u1}(T)$  behaves non-monotonically, and the high-temperature enhancement seen in Figs. 1 and 12 is to be considered as a finite-temperature reflection of the non-monotonic ground-state behavior in Fig. 2 rather than a reflection of the plain shift of the Curie temperature.

## V. CONCLUSIONS

Temperature dependence of MAE and magnetization for YCo<sub>5</sub> has been calculated from first principles within the assumption of local moments to give the qualitative agreement with the experimental data. We have proposed a way to improve the high-temperature coercivity by hole doping as was demonstrated by calculations with the filling controlled by the chemical potential manipulation. That can be implemented as applying the gate voltage for the case of a thin film sample, or alloying Co with some other elements to slightly reduce the valence electron number in the bulk. Since Co is an expensive element, replacing it with some cheaper ingredients with the improved coercivity in the practical operation temperature range is quite welcome. Thus we have proposed a way to improve the practical utility of YCo<sub>5</sub> from the perspective of the electronic structure and statistical physics.

## ACKNOWLEDGMENTS

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2013, and K. Hono for the overall push to the present project. Numerical computations were performed on “Minerva” at University of Warwick and SGI Altix at National Institute for Materials Science.

## Appendix A: Details of the calculations for $\text{YCo}_5$

### 1. Adjustment of the valence electron number

The overall calculation consists of two steps:

1. The first stage is to generate the grand potentials for each of the assumed local moment configurations by a scalar-relativistic KKR calculation. We do a spin-polarized calculation for  $\text{YCo}_5$  as parametrized by Vosko *et al.*<sup>45</sup> to reach the ferromagnetic ground state and yield the potentials for each local moment on the magnetic sublattices.
2. Then the second stage consists of solving the fully relativistic equation of state under the potentials generated by the electronic structure calculation in step 1 and the direction of magnetization is fixed to be  $\mathbf{n} = (1, 0, 1)$  with the motivation to address MAE using the torque-based formula in Eq. (15). Solutions are obtained so that Eqs. (3) and (5) are solved self-consistently to yield the result on MAE, magnetization, and the temperature.

In these KKR-based calculations we set the cutoff order in the expansion of spherical harmonics  $l_{\text{max}} = 3$  all through the above two steps.

In step 1, we follow the atomic-sphere approximation (ASA) considering the deformed shape of the unit cell to avoid the complication caused by overlapping muffin-tin spheres or too large a interstitial region. We have confirmed that this ASA construction is adequate from full potential (FP)-linear muffin tin orbital (LMTO) calculations where we find the ASA and FP-LMTO densities of states for the  $T = 0\text{K}$  cases to compare well.

Having difference in the treatment of the SOI between steps 1 and 2, adjustment of the Fermi level needs to be done to exactly fix the number of electrons, which is demonstrated in Fig. 13. The adjustment is done in the order of  $O(0.01)$  [Ry]  $\sim O(0.001)$  [eV]. We describe the practical procedure for this in Sec. A 3.

### 2. Achieving the self-consistency in DLM with multiple magnetic sublattices in the unit cell

Further iterations within DLM is taken for  $\text{YCo}_5$  to achieve the self-consistency of the calculation to incorporate the effects of multiple mean-fields that depend on the magnetic sublattice within the unit cell. The target material  $\text{YCo}_5$  has at least two different magnetic sublattices, Co(2c) and Co(3g), and the magnetic moments on them can be different from each other as was found in

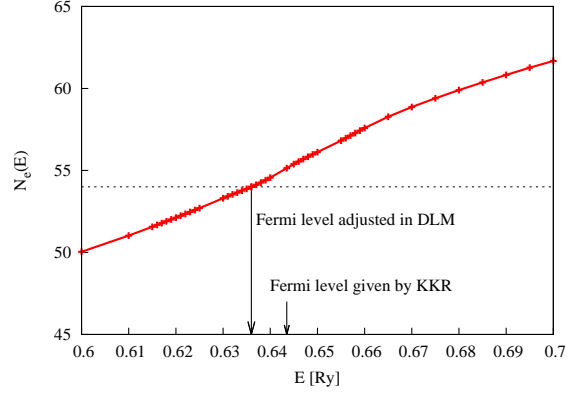


FIG. 13. (Color online) Calculated Fermi-level dependence of number of electrons for pure  $\text{YCo}_5$  near the ground state at  $m = 0.967$  [ $m = L(\beta h)$  with  $\beta h = 30$  as in Eq. (8)]. Equation of state is solved with the artificially given Fermi level to adjust the valence electron number exactly to the number as set up by the calculation.

the neutron scattering experiments<sup>26</sup>. The Weiss fields are obtained from Eqs. (3) and (5) solved self-consistently for the sites on the sub-lattice where there are local moments residing. We proceed as follows.

1. We start with a run with  $\beta h$  uniformly set over all of the local moments in the unit cell so that  $m_i = L(\beta h)$ . We refer to this run the zero-th round of the correction process.
2. With the output of the zero-th round, the input  $\beta h$ 's are re-set by using the output Weiss fields produced by the calculation from Eq (3).

$$\beta = (\beta h)_i / h_i \quad (\text{A1})$$

for all  $i$ 's. We note that the left-hand side, inverse temperature, is sublattice-independent. This imposes the self-consistency condition: we fix the input  $\beta h$  for Co(2c)-sublattice and adjust  $\beta h$  for other local moments that sit on the other sublattices according to the following relation:

$$\frac{(\beta h)_{\text{Co}(3g)}}{h_{\text{Co}(3g)}} \equiv \frac{(\beta h)_{\text{Co}(2c)}}{h_{\text{Co}(2c)}} \quad (\text{A2})$$

Using the output of the zero-th round for  $h$ 's, the input numbers for  $(\beta h)_{\text{Co}(3g)} = h_{\text{Co}(3g)} \times (\beta h)_{\text{Co}(2c)} / h_{\text{Co}(2c)}$  is used for the first round.

3. The process is iterated, making the 2nd, 3rd, ... rounds in the correction process, until the input  $S^1$  and the output  $S^1$ 's are identical within the numerical accuracy.

We have observed that for  $(\beta h)_{\text{Co}(2c)} \geq 10$  the convergence within 4 digits for  $h$ 's are quickly reached typically

after 3 iterations while for  $(\beta h)_{\text{Co}(2c)} \gtrsim 0.1$  the convergence takes around 10 iterations. The iteration can run into a limit cycle of a few periods beyond the 5th digit of  $\beta h$ . At such stage of the iteration, we regard that the convergence was reached within the numerical precision of 4 digits for  $S^1$  which we believe is sufficient.

### 3. Determining and varying the chemical potential in the DLM

In addressing magnetic hardness from first principles, magnetic anisotropy energy (MAE) needs to be calculated as precisely as possible. The behavior of the calculated MAE as a function of the valence electron number for  $\text{YCo}_5$  is shown in Fig. 2. This data is obtained in a series of DLM runs by artificially modulating the chemical potential near the ground state. Here the data near the ground state means the order parameter is fixed to be  $m = 0.967 = L(\beta h)$  with  $\beta h = 30$ . MAE has the strong dependence on the electron filling and only after fixing the electron number by a fine-tuning of the chemical potential in the DLM run, we can discuss the temperature dependence of MAE and magnetization from first principles: here we fix the valence electron number down to 4 digits by manual tuning the chemical potential. The fine-tuning follows the Newton's method in spirit, where we keep on shifting the chemical potential by  $\Delta\mu$  with

$$\Delta\mu = (N_{e,\text{desired}} - N_e)/n(\mu)$$

where  $N_e$  is the electron number given as  $N^{(\hat{n})}(\mu)$  and  $n(\mu)$  is the density of states at  $\mu$ , until the desired electron number  $N_{\text{desired}} = 54$  is numerically reached by iterating the calculations with the shifted chemical potential  $(\mu + \Delta\mu)$ . At this determined chemical potential and the electron number  $N_e = 54.00$  near the ground state, we find the low-temperature MAE to be 2.3 [meV/unit cell] as shown in Fig. 2. Using the experimental lattice constants<sup>26</sup>, this is equivalent to 4.4 [MJ/m<sup>3</sup>], which compares with the experimental result at liquid nitrogen temperature, 6.3 [MJ/m<sup>3</sup>]<sup>24</sup>, with an underestimate of 30%.

The above fine-tuning of the chemical potential also depends on the temperature. At each data point on the temperature axis, the valence electron number is fixed to be 54.00 within the numerical accuracy which practically

gives a temperature-dependent chemical potential from 0.63609 [Ry] at the lowest temperatures to 0.63357 [Ry] at the calculated highest temperature 965 [K].

### 4. Magnetization and local moments

The total magnetization, and its spin part, and the orbital part, is calculated to be 8.03 [ $\mu_B$ ], 7.44 [ $\mu_B$ ], and 0.59 [ $\mu_B$ ], respectively, as shown in Fig. 5 for low temperature. The calculated total magnetization amounts to  $J_s = 1.11$  [T] using the experimental lattice constants<sup>26</sup>.

	total	spin	orbital
Co(2c) (our calc.)	1.88	1.81	0.0667
Co(2c) (exp.)	1.77	1.31	0.46
Co(3g) (our calc. for Co(3)/Co(4))	1.59	1.44	0.150
(our calc. for Co(5))	1.60	1.44	0.164
Co(3g) (exp.)	1.72	1.44	0.28

TABLE III. Calculated magnitude of local moments in [ $\mu_B$ ]. Experimental numbers are taken from Ref. 26.

These results agree with the experimental numbers found in the literature of 7.99 [ $\mu_B$ ]<sup>26</sup> or  $J_s = 1.1$  [T]<sup>8</sup> within 0.5%.

However we note that this apparent excellent agreement is actually realized by a cancellation of overestimates and underestimates, which is revealed by inspecting each local moment's contribution to the total magnetization. At the fixed filling, our results are extracted as summarized in Table. III. Our calculated local moments on Co(2c)/Co(3g) sublattice are 1.88 and 1.59 [ $\mu_B$ ] which compare with the results from neutron experiments 1.77/1.72 [ $\mu_B$ ]<sup>26</sup> within 6-8%. Those polarizations are given by DLM in the fully relativistic calculation near the ground state, based on the potentials generated by the scalar-relativistic KKR calculation at  $T = 0$  which gave the polarization as 1.48 and 1.52 for Co(2c) and Co(3g), respectively. In the present local moment approach for  $\text{YCo}_5$  we have systematic underestimate of the orbital moments compared with the neutron measurements. However, the resolution of the neutron data into spin and orbital contribution also involves some subtlety<sup>46</sup>. The accurate *ab initio* estimation of the orbital contribution to the magnetization is left for future projects.

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